July 6, 2009

Mr. Roy Crossland
START Project Officer
U.S. Environmental Protection Agency, Region 7
901 North 5th Street
Kansas City, Kansas 66101

Subject: **Lead Trend Analysis – Evaluation by Individual Quadrants**
Herculaneum Lead Smelter, Herculaneum, Missouri
U.S. EPA Region 7 START 3, Contract No. EP-S7-06-01, Task Order No. 0021
Task Monitor: Bruce Morrison, On-Scene Coordinator

Dear Mr. Crossland:

Tetra Tech EM Inc. is submitting the attached updated Lead Trend Analysis at the Herculaneum Lead Smelter. Tetra Tech has updated the trend analysis to include the latest round of data obtained in April 2009. If you have any questions or comments, please contact the program manager at (816) 412-1754 or me at (816) 412-1762.

Sincerely,

David Homer, Ph.D.
Project Manager

Ted Faile, P.G.
START Program Manager

cc: Bruce Morrison, EPA
Ray Bienert, Tetra Tech

Enclosures

LEAD SOIL TREND ANALYSIS THROUGH APRIL 2009
EVALUATION BY INDIVIDUAL QUADRANT

HERCULANEUM LEAD SMELTER
HERCULANEUM, MISSOURI

CERCLIS ID: MOD006266373

Superfund Technical Assessment and Response Team (START) 3 Contract
Contract No. EP-S7-06-01, Task Order 0021

Prepared For:

U.S. Environmental Protection Agency
Region 7
Superfund Division
901 North 5th Street
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July 6, 2009

Prepared By:

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INTRODUCTION

Tetra Tech EM Inc. (Tetra Tech) was tasked by the U.S. Environmental Protection Agency (EPA) Region 7 Enforcement/Fund Lead Removal program to conduct a trend analysis of soil lead concentrations at selected locations within Herculaneum, Missouri (City). Specifically, the Tetra Tech Superfund Technical Assessment and Response Team (START) 3 was requested to review and analyze data that would enable EPA to determine if soil lead concentrations were increasing over time at a variety of locations within the City. Two tasks were identified: (1) perform a trend analysis for individual quadrants within each yard using the most current sampling data, and (2) estimate the range of monthly increase in lead concentrations for properties grouped into three categories based on distance from the smelter (less than or equal to 0.25 mile, 0.25 to 0.50 mile, and 0.50 to 0.75 mile). The assessment was conducted under the authority of the Comprehensive Environmental Response, Compensation, and Liability Act of 1980 and the Superfund Amendments and Reauthorization Act of 1986. The project was assigned under START Contract No. EP-S7-06-01, Task Order No. 0021.

Tetra Tech used analytical results provided by EPA (data set labeled "Recontamination") for measurements collected through round 26. Round 27 data were provided by the Doe Run Company (Doe Run); these data were added to the previous results. Both the EPA and Doe Run data sets included results from a number of residential properties. The data were collected from four different quadrants at each property, and additional data for several properties came from samples collected in driveway areas outside the quadrants. Lead sampling was conducted at each location at varying intervals from the time removal activities were completed in early 2002 (sampling round 6). Sampling was conducted monthly prior to 2003, quarterly from 2003 to 2004, and semi-annually after October 2005 (sampling round 22). This report includes results for sampling conducted between August 2002 (sampling round 7) and April 2009 (sampling round 27). Due to the sequence of removal activities, not all properties underwent the same number of sampling events; the number of events ranged from 9 to 21 events per quadrant for individual properties. At many locations, some intervals within the series were omitted because of weather or access restrictions. The lead concentrations were determined by use of a portable X-ray fluorescence (XRF) instrument. Samples were collected and analyzed in accordance with the quality assurance project plan (QAPP) dated September 11, 2001.

This document describes the methods used to evaluate changes in soil lead concentrations following the removal activities, and presents the results of this analysis.

Methods

Trend tests were conducted for each property using data collected from round 7 (August 2002) through round 27 (April 2009). The nonparametric Mann-Kendall test was used to evaluate temporal trends for each quadrant sampled at the individual properties. The Mann-Kendall test is a widely used statistical test for detecting monotonic trends (that is, trends that are either increasing or decreasing) in time-series of data (Gilbert 1987, Helsel and Hirsch 1992, Gibbons 1994). Because the Mann-Kendall test uses only the relative magnitude of the data rather than their measured values, it has a number of desirable properties: the data need not be normally distributed; and the test is not significantly affected by outliers, missing data, or censored (nondetect or below the detection limit) data. Censored data are treated in the Mann-Kendall test by setting all nondetect results to a concentration slightly below the minimum detected concentration. A minimum of four sampling events are required to perform this test, so properties with fewer than four rounds of sampling were not evaluated. Properties not sampled during round 27 were also excluded from the trend analysis.

For all properties where at least one quadrant showed a significant increasing trend based on the Mann-Kendall test, regression analysis was performed to estimate the monthly rate of increase in lead concentrations. This analysis was performed to provide rough estimates of the range of potential increase in lead concentrations for properties grouped according to distance from the smelter. Three distance categories were evaluated: less than or equal to 0.25 mile, 0.25 to 0.50 mile, and 0.50 to 0.75 mile. Because the purpose of this analysis was to provide only rough estimates of the rate of change in lead concentration, regression was performed on the data in original units (i.e., untransformed data). It should be noted that certain evaluation methods and diagnostic tools available to optimize and improve the reliability of linear regression results (e.g., evaluation of different transformations of the data, verification of model assumptions, and evaluation of outliers) were not used in this analysis. For this reason, caution should be exercised in interpreting forecasted estimates of the rate of increase in lead concentrations.

For quadrants with detected data only, ordinary least squares (OLS) linear regression analysis was used. For quadrants with one or more censored results, a parametric maximum likelihood estimation (MLE) approach was used, following Helsel (2005). MLE methods are used increasingly in environmental assessment work, given the increased speed of modern personal computers and the enhanced capabilities of many commercial and public domain statistical software packages. As described in Helsel (2005), MLE regression techniques can be implemented using commercial software with capabilities for performing parametric survival analysis on interval-censored data (MLE regression for left-censored data is also referred to as “Tobit analysis” in the technical literature). MLE methods recognize each censored datum as an interval, bounded by zero at the lower limit and the detection or reporting limit at the upper limit. Application of OLS regression with censored data is contraindicated, as it requires substitution of an assumed value (typically zero, the detection limit, or one half the detection limit) for each censored datum, resulting in biased estimates for the regression parameters.

Results

Temporal trends in lead concentrations for 13 properties are summarized in Table 1 and Figure 1. All 13 properties had at least one quadrant that showed a statistically significant increasing trend. No statistically significant decreasing trends were identified for any properties. Four properties had increasing lead concentrations in all four quadrants: house numbers 9, 18, 19, and 24. Three properties had increasing lead concentrations in three quadrants: house numbers 3, 10 (only three quadrants evaluated), and 13 (only three quadrants evaluated). Three properties had increasing lead concentrations in two quadrants: house numbers 5 (only two quadrants evaluated), 7, and 76 (only two quadrants evaluated). Three properties had increasing lead concentrations in one quadrant: house numbers 16 (only 1 quadrant evaluated), 103, and 104 (only three quadrants evaluated). All trend results are depicted graphically in Figure 1 for individual properties ordered by increasing distance from the smelter. Censored results are plotted at a concentration slightly below the minimum detected result to reflect the rank order of the data evaluated in the Mann-Kendall test.

Trend results reported for soil lead concentrations through sampling round 27 were similar to those reported during the last quarterly period, with the following exceptions. Selected quadrants from three properties that did not show increasing trends in round 26 (quadrants 2 and 4 from house number 13; quadrants 1 and 4 from house number 10; and quadrant 1 from house number 103) showed statistically significant increasing trends in round 27. Note that quadrant 2 from house number 10 was evaluated in round 26 but not round 27, and houses 5, 16, and, 19 were evaluated in round 27 but were not evaluated in round 26.

Table 2 shows results of OLS and MLE regression analysis performed on properties that showed a significant increasing trend in lead concentration in at least one quadrant. The slope, intercept, standard error of the slope, and two-sided 95 percent confidence intervals for the slope estimates were calculated for 30 quadrants from 10 properties. Ranges for the monthly rates of increase in lead were 1.3 to 22 milligrams (mg)/month for properties located less than or equal to 0.25 mile from the smelter, 0.6 to 3 mg/month for distances between 0.25 and 0.50 mile, and 0.3 to 6 mg/month for distances between 0.50 and 0.75 mile from the smelter. The upper 95 percent confidence limit (UCL) for the monthly rate of increase was also evaluated to estimate maximum potential rates of increase. Because of the variability in the individual estimates, the 50^{th}, 75^{th}, and 90^{th} percentiles of the distribution of the individual UCLs within each distance category are also reported in Table 2. The 75^{th} and 90^{th} (in parentheses) percentile values for the monthly rate of increase for the properties located less than or equal to 0.25 mile from the smelter are 9 (42) mg/month, between 0.25 and 0.50 mile are 4 (5) mg/month, and between 0.50 and 0.75 mile from the smelter are 3 (6) mg/month. Caution interpreting these results is necessary because these are considered rough estimates only—no attempt was made to evaluate the validity of the regression model assumptions or the uncertainty associated with the predicted rates of increase.

References:

Gibbons, R. D. 1994. *Statistical Methods for Groundwater Monitoring*. John Wiley & Sons, Inc. New York, New York.

Gilbert, R. O. 1987. *Statistical Methods in Environmental Pollution Monitoring*. John Wiley & Sons, Inc. New York, New York.

Helsel, D. 2005. *Nondetects and Data Analysis: Statistics for Censored Environmental Data*. John Wiley & Sons, Inc. New York, NY.

Helsel, D. R. and R. M. Hirsh. 1992. *Statistical Methods in Water Resources*. Elsevier. New York, New York.

TABLE 1
RESULTS OF STATISTICAL TESTING FOR MONOTONIC TRENDS (MANN-KENDALL TEST) IN LEAD CONCENTRATION
INDIVIDUAL QUADRANTS FOR SAMPLING ROUNDS 7 THROUGH 27
HERCULANEUM LEAD SMELTER SITE - HERCULANEUM, MISSOURI

Distance From Smelter[1]	House Number	Address	Quadrant	Number of Sampling Events[2]	Number of Detected Samples	Sampling Event		Mann-Kendall Test Statistic[3] (S)	Probability > S	Trend Significant?[4] (Yes/No)	Direction of Trend
						First	Last				
0.10	76		Q1	14	14	10/30/2003	04/26/2009	68	0.000	**Yes**	**Increasing**
			Q2	14	14	10/30/2003	04/26/2009	55	0.003	**Yes**	**Increasing**
0.25	5		Q1	19	16	08/26/2002	04/25/2009	131	0.000	**Yes**	**Increasing**
			Q4	19	19	08/26/2002	04/25/2009	113	0.000	**Yes**	**Increasing**
0.25	24		Q1	17	17	11/07/2002	04/25/2009	78	0.001	**Yes**	**Increasing**
			Q2	17	17	11/07/2002	04/25/2009	104	0.000	**Yes**	**Increasing**
			Q3	17	17	11/07/2002	04/25/2009	90	0.000	**Yes**	**Increasing**
			Q4	17	16	11/07/2002	04/25/2009	79	0.001	**Yes**	**Increasing**
0.40	13		Q1	10	10	08/23/2002	04/25/2009	27	0.008	**Yes**	**Increasing**
			Q2	10	10	08/23/2002	04/25/2009	21	0.036	**Yes**	**Increasing**
			Q4	10	9	08/23/2002	04/25/2009	23	0.023	**Yes**	**Increasing**
0.50	16		Q2	15	9	09/16/2002	04/26/2009	61	0.002	**Yes**	**Increasing**
0.50	19		Q1	18	17	08/22/2002	04/25/2009	70	0.007	**Yes**	**Increasing**
			Q2	18	15	08/22/2002	04/25/2009	84	0.001	**Yes**	**Increasing**
			Q3	18	15	08/22/2002	04/25/2009	70	0.006	**Yes**	**Increasing**
			Q4	18	17	08/22/2002	04/25/2009	83	0.002	**Yes**	**Increasing**
0.54	9		Q1	20	20	08/22/2002	04/25/2009	109	0.000	**Yes**	**Increasing**
			Q2	20	20	08/22/2002	04/25/2009	102	0.001	**Yes**	**Increasing**
			Q3	20	20	08/22/2002	04/25/2009	112	0.000	**Yes**	**Increasing**
			Q4	20	19	08/22/2002	04/25/2009	103	0.001	**Yes**	**Increasing**
0.60	18		Q1	21	21	08/23/2002	04/25/2009	112	0.001	**Yes**	**Increasing**
			Q2	21	20	08/23/2002	04/25/2009	103	0.002	**Yes**	**Increasing**
			Q3	21	21	08/23/2002	04/25/2009	125	0.000	**Yes**	**Increasing**
			Q4	21	21	08/23/2002	04/25/2009	126	0.000	**Yes**	**Increasing**
0.75	3		Q1	21	18	08/23/2002	04/26/2009	39	0.103	No	N/A
			Q2	21	19	08/23/2002	04/26/2009	104	0.002	**Yes**	**Increasing**
			Q3	21	20	08/23/2002	04/26/2009	93	0.004	**Yes**	**Increasing**
			Q4	21	20	08/23/2002	04/26/2009	133	0.000	**Yes**	**Increasing**
	10		Q1	9	7	08/22/2002	04/25/2009	17	0.049	**Yes**	**Increasing**
			Q3	9	5	08/22/2002	04/25/2009	27	0.002	**Yes**	**Increasing**
			Q4	9	4	08/22/2002	04/25/2009	21	0.017	**Yes**	**Increasing**

Distance From Smelter[1]	House Number	Address	Quadrant	Number of Sampling Events[2]	Number of Detected Samples	Sampling Event		Mann-Kendall Test Statistic[3] (S)	Probability > S	Trend Significant?[4] (Yes/No)	Direction of Trend
						First	Last				
0.79	103		Q1	8	4	03/28/2005	04/25/2009	16	0.031	**Yes**	**Increasing**
			Q2	8	4	03/28/2005	04/25/2009	6	0.274	No	N/A
			Q3	8	4	03/28/2005	04/25/2009	12	0.089	No	N/A
			Q4	8	6	03/28/2005	04/25/2009	1	0.500	No	N/A
0.80	7		Q1	21	21	08/23/2002	04/24/2009	39	0.103	No	N/A
			Q2	21	18	08/23/2002	04/24/2009	95	0.004	**Yes**	**Increasing**
			Q3	21	17	08/23/2002	04/24/2009	47	0.075	No	N/A
			Q4	21	16	08/23/2002	04/24/2009	98	0.003	**Yes**	**Increasing**
1.00	104		Q1	8	6	03/28/2005	04/24/2009	3	0.406	No	N/A
			Q2	8	6	03/28/2005	04/24/2009	18	0.016	**Yes**	**Increasing**
			Q4	8	4	03/28/2005	04/24/2009	10	0.138	No	N/A

Notes:

1 Properties are ordered as a function of increasing distance from the smelter.

2 Trend tests were not conducted for properties with fewer than four rounds of sampling, or for properties that were not sampled during round 27.

3 All censored (nondetect) measurements were set equal to a concentration slightly lower than the minimum detected result.

4 Monotonic trends are significant for probabilities less than or equal to 0.05; significant negative values for the Mann-Kendall test statistic A1indicate that trends are decreasing. Significant positive values for the Mann-Kendall test statistic indicate that trends are increasing.

N/A No significant trend identified.

TABLE 2
RESULTS OF LINEAR REGRESSION ANALYSIS FOR ALL QUADRANTS SHOWING A SIGNIFICANT INCREASING MANN-KENDALL TREND TEST RESULT

Distance From Smelter (Miles)	House Number	Quadrant	Number of Sampling Events	Regression Coefficients for Days Versus Concentration			Monthly Increase (mg/kg-month)	95 Percent Confidence Limits for Monthly Increase in Lead Concentrations[1]		Percentiles for the Distribution of Estimated UCLs within Each Distance Group		
				Intercept	Slope	S.E. (Slope)		LCL	UCL	50	75	90
Less than or Equal to 0.25	76	Q1	14	-4.90	0.21	0.04	6.32	3.48	9.16	4.40	9.09	42.05
		Q2	14	96.33	0.09	0.04	2.71	0.40	5.02			
	5	Q1	19	43.59	0.09	0.02	2.77	1.76	3.78			
		Q4	19	65.82	0.19	0.05	5.86	2.83	8.88			
	24	Q1	17	128.68	0.07	0.02	2.19	1.06	3.32			
		Q2	17	-277.65	0.72	0.31	21.78	1.51	42.05			
		Q3	17	50.10	0.07	0.02	2.12	1.09	3.16			
		Q4	17	70.42	0.04	0.01	1.32	0.56	2.08			
0.25 to 0.50	13	Q1	10	153.30	0.07	0.02	2.01	0.53	3.49	3.31	3.75	4.73
		Q2	10	144.14	0.09	0.03	2.70	0.67	4.73			
		Q4	10	137.16	0.04	0.01	1.24	0.36	2.12			
	16	Q2	15	71.09	0.07	0.02	2.20	0.68	3.72			
	19	Q1	18	55.05	0.04	0.01	1.22	0.63	1.81			
		Q2	18	24.34	0.10	0.01	2.89	2.01	3.77			
		Q3	18	50.14	0.02	0.01	0.64	-0.10	1.38			
		Q4	18	50.88	0.07	0.01	2.20	1.25	3.14			
0.50 to 0.75	9	Q1	20	63.05	0.05	0.01	1.54	0.83	2.25	2.01	2.79	6.27
		Q2	20	57.03	0.10	0.02	2.95	1.54	4.36			
		Q3	20	127.37	0.18	0.04	5.55	2.91	8.19			
		Q4	20	97.36	0.08	0.02	2.36	1.19	3.53			
	18	Q1	21	75.28	0.05	0.01	1.44	0.57	2.32			
		Q2	21	53.59	0.05	0.01	1.48	0.94	2.02			
		Q3	21	72.68	0.03	0.01	0.77	0.45	1.09			
		Q4	21	65.89	0.04	0.01	1.20	0.81	1.59			
	3	Q2	21	59.34	0.03	0.01	0.82	0.30	1.34			
		Q3	21	46.46	0.03	0.01	0.86	0.42	1.29			
		Q4	21	48.06	0.04	0.01	1.11	0.71	1.50			
	10	Q1	9	39.48	0.05	0.01	1.51	0.47	2.55			
		Q3	9	25.40	0.04	0.01	1.20	0.40	2.00			
		Q4	9	37.97	0.01	0.00	0.27	0.02	0.53			

Notes:

Houses within each distance group are sorted by increasing distance from the smelter.
Results are for sampling rounds 7 through 27.

kg Kilogram
LCL Lower confidence limit
mg Milligram
MLE Maximum likelihood estimate
ND Nondetect
OLS Ordinary least squares
S.E. Standard error of estimate
UCL Upper confidence limit

OLS regression was used for cases where all results were detected. Parametric MLE regression with a normal model assumption was used in all cases where one or more measurements were reported as below the detection limit (that is, "ND") following Helsel (2005). All analyses were performed on the data in original units.

[1] Monthly increases are not statistically significant at the 95 percent confidence level if the confidence interval includes zero.

Helsel, D. 2005. *Nondetects and Data Analysis: Statistics for Censored Environmental Data* . John Wiley & Sons, Inc., New York, NY. 250 pages.